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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US97/22069 <b>(22) International Filing Date:</b> 5 December 1997 (05.12.97) <b>(30) Priority Data:</b> 08/761,626 5 December 1996 (05.12.96) US <b>(71) Applicant:</b> EXXON CHEMICAL PATENTS INC. [US/US]; 5200 Bayway Drive, Baytown, TX 77520-5200 (US). <b>(72) Inventor:</b> THOMPSON, Francis, Murphy; 12 Shepherds Lane, Whitehouse Station, NJ 08889 (US). <b>(74) Agents:</b> MILLER, Douglas, W. et al.; Exxon Chemical Company, P.O. Box 2149, Baytown, TX 77522-2149 (US).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, HU, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, ARIPO patent (GH, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
<b>(54) Title:</b> DISPERSIBLE FILM  <b>(57) Abstract</b>  Compounding ingredients for unvulcanized rubber, or bales of unvulcanized rubber are packaged in filler reinforced film or bags made from an ethylene $\alpha$ -olefin copolymer having a Vicat softening point below 90 °C and a melting point below 100 °C, and optionally a second ethylene copolymer is added to improve processability of the first ethylene $\alpha$ -olefin copolymer. The second ethylene copolymer will contain more long-chain branches than the first ethylene copolymer. The reinforcing filler is selected from talc, especially talc with a high aspect ratio, silica, or other mineral filler, or carbon black, having a strengthening effect on the film. The film will display relatively high 1 % secant modulus and a relatively high Elmendorf tear, for a film having a low Vicat softening point.		

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## DISPERSIBLE FILM

### TECHNICAL FIELD

This invention relates to a polymer film used to make compounding bags and rubber bale wrap for rubber mixing, but also extends to polymer films used to wrap or contain ingredients for addition to most polymer/additive mixes requiring a relatively low process temperature at which temperature the polymer film, bag, or over-wrap based on the polymer film must disperse into the mix.

### BACKGROUND

In the manufacture of rubber goods, elastomers and additives (generally compounding ingredients) are combined in a mixer to form a substantially homogeneous mixture (or rubber compound) which is then formed to the desired finished product shape. This shaped mixture may then be cured, or cross linked. The economics of making rubber induce the manufacturer to make ever more rapid mixes and ever more rapid cures in order to remain competitive. Rapid curing usually implies lower temperature mixing which in turn limits mixing temperature because the mix temperature must not exceed the activation temperature of the curatives. These trends encourage the use of rubber bale wrap and/or compounding bags using films that melt or soften at ever lower temperatures to facilitate rapid dispersion of the film in the rubber/additive matrix. Compounding bags are used to hold a variety of additives including, but not limited to curatives such as sulfur, and primary and secondary accelerators; fillers both reinforcing and non-reinforcing; plasticizers such as oil and waxes; tackifiers; and specialty chemical additives used as coupling agents and stabilizers for heat, ozone, UV light, and the like. Other polymer mixing and or compounding operations are analogous to mixing elastomers in the desire to carry bagged compounding ingredients for addition to a mixing process which requires an easily dispersed bag.

The use of pre-weighed ingredients has many advantages over the direct addition by "scoop and shovel," 1) improved housekeeping, 2) improved industrial hygiene, 3) loss control, 4) accuracy and consistency for improved quality, 5) reduced labor costs. The actual weighing/measuring can be done remotely or very

near the point of addition to the mixer. It can be a relatively simple manual operation or a more elaborate automated form-fill-seal operation. In any case, the result is a measured quantity of wrapped or bagged material suitable for direct addition to the mixer.

5 Polyethylene film has been used for compounding ingredient bags and/or rubber bale wraps because of its generally low cost, ability to be formed, filled and sealed, and toughness. Very early, the limits of polyethylene became apparent and restricted its use. The relatively high melt temperature and Vicat softening point of the low density polyethylene restricted it to relatively high temperature mixes.  
10 Ethylene copolymers having lower melting and Vicat softening points were introduced such as ethylene vinyl acetate (EVA) copolymers or other low melting point ethylene polar comonomer copolymers. More recently metallocene catalyzed low or very low density polyethylene (generally m-polyethylene) and blends thereof have also been used and/or discussed. Additives, such as low melting waxes, have  
15 also been employed to lower these temperatures. However, these solutions also encountered limits as the need for ever lower temperatures developed.

Various problems associated with using polyethylene and/or EVA copolymer films used for bale wrap and compounding bags have been mentioned here and in US 5,500,260 and incorporated herein by reference for purposes of US  
20 patent practice. In particular, this patent suggests the need for a film to have a Vicat softening point as low as or below 82° C (also disclosed in US 5,145,747) to provide adequate dispersion of the film in an elastomer mix.

There is now a need to obtain adequate film dispersion at even lower temperatures, particularly requiring a film with a melt point as low or lower than  
25 75° C for certain rubber mixes (and a Vicat of as low as or lower than 55° C). Requirements for even lower Vicat softening points are being developed with some industry requirements now as low as 48° C Vicat softening point.

However, as the Vicat softening point and melting point of a polyethylene based film is reduced, the strength properties of films based on such polyethylenes  
30 or polyethylene blends having such melt/Vicat combinations, generally decrease, often dramatically decreasing, creating practical problems with handling, shipping,

and storing unused film and bags and then problems with handling film and bags after filling. A film with such low Vicat softening point as for instance 55° C becomes very soft, stretchy, and tacky and will generally have low resistance to tearing making it difficult to handle in either wrap or bag making operations or further downstream in filling, storage and handling. To compensate for these low strength properties, the film and/or bags must be either handled somewhat delicately to prevent damage or greater thickness of film must be used to compensate for the low strength. Both of these compensations are commercially impractical. Greater thickness are both less economical and introduce more film into a rubber mix that requires dispersion. Anti-block additives and slip (generally erucamide) are used to overcome tack, but especially the slip may make the bag slippery especially at slip levels high enough to mitigate sticky films created from such low melting or softening polyethylenes. Such slippery films are difficult to handle. The use of anti-block additives is common in the film industry at levels as low as 1,000 ppm (0.1 wt%) to levels of as high as 5 wt. %. The use of calcium carbonate (CaCO<sub>3</sub>) as a filler in such films is known. However, while CaCO<sub>3</sub> improves some properties of low melting and/or low Vicat softening point films, still more improvement is desired.

A suggested solution to these problems suggested in Patent Number PCT/US94/09988, is the use of polyethylenes that form stronger films to blend or co-extrude with these low melt materials. Unfortunately, these polyethylenes that form stronger films generally have higher melting points and thus detract from overall dispersability of films including them.

There is a commercial need therefore for a polyethylene film, preferably a polyethylene film that has a relatively low melting point as well as a relatively low Vicat softening point, that will additionally have physical properties (strength) sufficient to allow normal shipping and handling without substantial breakage.

### SUMMARY

Embodiments of my invention overcome the generally lower strength of lower melting point polyethylene films by reinforcing the films with a suitable filler, without substantially effecting (raising) the melt point and/or Vicat softening point.

Fillers such as talc, more preferably talc with a high aspect ratio, and most preferred are such fillers or combinations thereof that are surface treated. Silica, carbon black and other fillers having a reinforcing effect may also be used. The use of a reinforcing filler has several beneficial effects. The filler or filler combination can make the film polyethylene matrix stiffer and tougher as measured by secant modulus, puncture and tear resistance of the films. This improved stiffness and toughness allows the use of lower thickness film which reduces both the amount of the polyethylene added to the intended elastomeric matrix and also reduces the amount of film material that needs to be dispersed. The reinforcing filler also appears to modify the rheology of the polyethylene to make film extrusion more economical by reducing power requirements and increasing extrusion rate. The desirable effects of filler depend on the amount of reinforcing filler added as well as the type, but the desirable effects generally become effective in the range of from 5%-50% filler by weight, preferably in the range of from 10-30 percent by weight based on the total weight of the polyethylene/filler combination. Such levels are above and different from the above mentioned anti-block materials and their levels.

With embodiments of my invention, I have found that the above-mentioned disadvantages associated with prior solutions can be minimized or eliminated by the use of a film made from at least one filler reinforced low density polyethylene resin, where the resin from which the film is made has a Vicat softening point up to 90° C and preferably a density up to 0.915 g/cm<sup>3</sup>. In addition, the film (in a softened or melted state) is generally compatible with rubber compounds. A film having a melt point temperature to achieve such compatibility will generally also allow shorter mixing times for the rubber being compounded, therefore providing for greater productivity. The film's low melting temperature (below 100° C), substantially insures fluxing into the rubber compound mix during even abbreviated mixing cycles. A melted film facilitates dispersion into a rubber mix. Since the temperature of a particular mix is generally a function of mixing time, a lower melting and/or softening film can be dispersed in a shorter time. These lower melting/softening point films enable the rubber manufacturer to achieve shorter mixing times.

In one aspect of my invention, a polyethylene film, formed from a filler reinforced ethylene- $\alpha$ -olefin copolymer, is provided wherein such film has:

- a) a polyethylene resin density, before reinforcement, and before film formation, in the range of from 0.85- 0.915 g/cm<sup>3</sup>;
- b) a polyethylene melt index, before reinforcement, in the range from 0.1- 10 grams per 10 minutes;
- c) a reinforcing filler content in the range of from 5-50 weight percent based on the total weight of the polyethylene filler combination;
- d) a melting point of the filled reinforced film not exceeding 100° C.; and
- e) a Vicat softening point of the filled reinforced film not exceeding 90° C.

In another embodiment, a second ethylene copolymer may be added to improve the melt processability of the first ethylene copolymer. The second ethylene copolymer may be a copolymer of ethylene and an ethylinically unsaturated ester of a carboxylic acid. The second ethylene copolymer is preferably any polymer with sufficient branching to contribute to the melt processability of the first ethylene copolymer (generally, but not necessarily so called "high-pressure" polyethylene). In this embodiment, the first ethylene copolymer, an ethylene  $\alpha$ -olefin copolymer, is present in the range of from 75-99 percent by weight of the polyethylene portion of the polyethylene/filler blend weight. The second ethylene copolymer is present in the range of from 25-1 weight percent based on polyethylene blend weight. By polyethylene blend weight, I intend that such a polyethylene blend will then form the basis for addition of filler, as stated above, accordingly, a polyethylene blend within the limits stated above will then have filler added, to form the polyethylene/filler blend.

#### **DETAILED DESCRIPTION**

Certain embodiments of my invention concern certain polyethylene films, their production and applications based on such films. These films have unique properties which make them particularly well suited for use in certain compounding or mixing operations. These films have combinations of properties rendering them superior to films previously available for many such compounding or manufacturing operations. Following is a detailed description of certain preferred films based on



polyethylene/filler or polyethylene blend filler combinations within the scope of my invention, preferred methods of producing these films, and preferred applications of these films. Those skilled in the art will appreciate that numerous modifications to these preferred embodiments can be made without departing from the scope of the invention. For example, though the properties of films based on polyethylene and/or polyethylene blend/filler combinations are exemplified in elastomer compounding applications, they have numerous other uses such as plastics compounding. To the extent my description is specific, this is solely for the purpose of illustrating preferred embodiments of our invention and should not be taken as limiting my invention to these specific embodiments.

Various Test Methods values given in the text and claims are determined as follows:

Melt index (MI): ASTM D 1238 Condition E 190° C, 2.16 kg mass; expressed in g/10 min.

Vicat softening point: ASTM-1525 with a 1000 g weight. (° C)

Melting point: Differential Scanning Calorimeter (DSC) second melting curve melting peak in degrees centigrade. (ASTM D-3417)

STIFFNESS (MODULUS)	ASTM	<u>D-882</u>	(expressed in pounds per square inch)
TEAR (ELMENDORF)	ASTM	<u>D-1922</u>	(expressed in grams or grams/mil)

The film of the present invention for use as a rubber bale wrap or compounding bag, has the following properties: good seal strength sufficient to effect seals to contain compounding ingredients, a low melting point, good sealability including sufficient hot tack for use in form-fill-and seal applications, a low Vicat softening point, improved resistance to tearing and improved stiffness level as measured by secant modulus in the filler polyethylene film combinations. The low Vicat softening point and the low melting point improve the ability of the film to be incorporated into a substantially homogeneous mixture of rubber and/or rubber compounding ingredients to form generally homogeneous rubber compounds.

The film of the present invention for use as rubber bale wrap and compounding bags may be made from a single first ethylene  $\alpha$ -olefin copolymer or a

blend of the first ethylene  $\alpha$ -olefin copolymer with one or more other ethylene  $\alpha$ -olefin copolymers, or with ethylene homopolymers, as long as the Vicat softening point is acceptable.

These ethylene  $\alpha$ -olefin copolymers or blends thereof are combined with a reinforcing filler using a means of mixing (well known to those of ordinary skill) to provide good filler dispersion resulting in the reinforcement of films based on this polymer filler mixture. This means of mixing includes, but is not limited to, single and dual screw mixing extruders as well as continuous mixers. Batch mixers would be expected to provide appropriate mixing and reinforcement as well.

The film made from the ethylene  $\alpha$ -olefin polyethylene/filler mixture has a Vicat softening point below 90° C and a melting point below 110° C. Preferably, the film has a Vicat softening point below 80° C, more preferably below 70° C, most preferably below 60° C and a DSC melting point below 100° C, preferably below 90° C, more preferably below 80° C.

In an embodiment of the present invention, a film is made from the mixture of polyethylene and filler for either bale wrapping or for making compounding bags. In either case, the film must have softening and melting characteristics as described supra that permit its inclusion into the rubber compounding process, and such inclusion will result in a substantially homogeneous compounded elastomer blend.

#### The Polyethylenes

The films of certain embodiments of my invention may be made from a reinforced mixture of an ethylene  $\alpha$ -olefin copolymer (first ethylene copolymer). This first ethylene copolymer has a density in the range of from 0.85-0.915 g/cm<sup>3</sup>, a Vicat softening point less than 90° C and a differential scanning calorimeter (DSC) second melting point (melting point) not exceeding 100° C. The  $\alpha$ -olefin utilized to make the first ethylene copolymer is selected from one or more of propylene, butene-1, 4-methyl-1-pentene, pentene-1, hexene-1, octene-1 decene-1 and mixtures thereof. I also intend that more than one such ethylene  $\alpha$ -olefin copolymer such combination include, but are not limited to copolymers such as ethylene propylene; ethylene, butene-1; ethylene, hexane-1; ethylene, pentene-1; ethylene 4-methyl-1-pentene;

ethylene, octene-1; ethylene, propylene; butene-1; ethylene, propylene, hexene-1; ethylene propylene, pentene-1; ethylene propylene, octene-1; ethylene, and the like may be used as the polyethylene portion of the polyethylene/filler blend, as long as the melting and/or Vicat softening point remains within the preferred levels

5           The first ethylene copolymer may be made by a number of processes, including low pressure, gas phase, fluidized bed, slurry or solution processes. The catalysts used for the polymerization are generally of the metallocene-alumoxane, metallocene-ionizing activator, or conventional Ziegler-Natta types. Such catalysts are well known. Thus, useful catalysts are those disclosed in EP 1229368, U. S. Patents  
10       numbers 5,026,798 and 5,198,401 incorporated herein by reference for purposes of US patent practice.

          Optionally, one or more second ethylene copolymers may be blended into the first ethylene  $\alpha$ -olefin copolymer or copolymers. The second ethylene copolymer is preferably not used primarily for depressing either the Vicat softening point or the  
15       melting point of the first ethylene copolymer, although such use is not precluded. Generally, the purpose of the second ethylene copolymer may be to improve melt processability (higher melt strength) of the polyethylene blend over the processability of the first ethylene copolymer alone. The second ethylene copolymer is generally a  
20       molecule containing branching typical of those produced in high-pressure, free radical processes well known to those of ordinary skill. In another embodiment of the present invention, the second ethylene polymer is a polyethylene homopolymer or preferably an ethylene copolymer of ethylene and at least one ethylinically unsaturated  
25       carboxylic acid ester. Preferred ethylinically unsaturated acrylic acid esters include, for example, vinyl acetate, methyl acrylate, butyl acrylate, ethyl acrylate and combinations thereof. A preferred ester monomer is vinyl acetate. These  
30       comonomers are present in the second ethylene polymer within a range of from 1-35 weight percent, preferably from 1-30 weight percent of the unsaturated acrylic acid ester, based on the total weight of the second ethylene copolymer. Blends of these ethylene copolymers can also be used.

          The second ethylene copolymers are chosen, in general, primarily based on their ability to enhance processability of the first ethylene polymer and their melting

points and/or Vicat softening points. That is, the melting points and/or Vicat softening points of such polymers should not have a substantial deleterious effect on these same parameters of the first ethylene copolymer, and the film made therefrom at the level of inclusion in the blends, but will mitigate processing debits sometimes found in the first ethylene copolymers. In general, a 10% improvement in bubble stability or neck-in over those of the first ethylene copolymer would be desirable as a result of inclusion of the second ethylene copolymer or copolymers. By substantial deleterious effect, I intend that inclusion of a second ethylene copolymer or copolymers would not raise the melting point and/or softening point above the discussed preferred levels.

When the second ethylene copolymer is included in the manufacture of film based on the ethylene  $\alpha$ -olefin copolymers described above, it is present in the range of from 25-1 weight percent based on the total polyethylene weight of the blend. The first ethylene  $\alpha$ -olefin copolymer is present in the range of from 75- 99 weight percent, based on the total polymer weight of the blend.

In a preferred embodiment, the first ethylene  $\alpha$ -olefin copolymer is present in the range of from 80-95 weight percent based on the total polymer weight of the polymer blend. The second ethylene copolymer is present in the range of from 20-5 weight percent based on the total polymer weight of the polymer blend. More preferred is a first ethylene  $\alpha$ -olefin copolymer of from 85-95 weight percent and a second ethylene copolymer from 5-15 weight percent. These weight percents are based on the total weight of the polyethylene portion of the polyethylene/filler blends.

Additionally, the reinforced film will further exhibit a 1% secant modulus machine direction (MD) of greater than 6,000 psi, preferably greater than 7,000 psi, more preferably greater than 8,000 psi, most preferably greater than 10,000 psi. Additionally, the film will exhibit a 1% secant modulus transverse direction (TD) greater than 6,000 psi, preferably greater than 7,000 psi, more preferably greater than 8,000 psi, most preferably greater than 9,000 psi.

The film will exhibit an Elmendorf tear as follows (g/mil):

	<u>MD</u>	<u>TD</u>
	>150	>140
preferred	>160	>160
more preferred		>200
most preferred		>250

It should be understood that the film produced from either a first ethylene copolymer, or a blend of at least two different ethylene  $\alpha$ -olefin copolymers (e.g. a first ethylene  $\alpha$ -olefin copolymer and a one or more other ethylene  $\alpha$ -olefin copolymers) may be reinforced with filler and used for film manufacture without the use of the second ethylene copolymer such as an ethylene vinyl acetate copolymer. However, in either the case where the first ethylene  $\alpha$ -olefin copolymer is reinforced and used alone, or in the case where it is blended with at least an other ethylene  $\alpha$ -olefin copolymer, it is understood that the Vicat softening point of the resulting film which may include filler does not exceed 90° C, and the melting point of the film does not exceed 100° C.

Also contemplated are films made from ethylinically unsaturated carboxylic acid ester copolymers with the disclosed fillers, in the substantial absence of an ethylene  $\alpha$ -olefin copolymer. However, it is also contemplated that such films will still exhibit the Vicat softening point and melting point upper limits disclosed, and preferably one or more of the secant modulus and/or tear strength limits.

Further contemplated are blends of the ethylinically unsaturated carboxylic acid ester copolymers as the majority component in the polyethylene portion of the polyethylene /filler blends used for the films of certain embodiments of the present invention.

Accordingly, such embodiments might include the following:

	Ethylene ethylinically unsaturated carboxylic acid ester copolymer	Ethylene $\alpha$ -olefin copolymer
Preferred	75-100	25-0
More preferred	85-95	5-15

The area of blends from 75% weight to 25% weight ethylene  $\alpha$ -olefin copolymer with the balance as the ethylene ethylinically unsaturated carboxylic acid ester copolymer are not excluded.

In all cases the polyethylene portion, whether a blend, or a single polyethylene, then makes up the polyethylene portion of the polyethylene/filler mixture. The mixture, when converted into film should display one or more of the described Vicat softening point, melting temperature, 1% secant modulus and/or Elmendorf tear.

#### The Filler/Reinforcement

It should be understood that the reinforcement of the polyethylenes (one or more of both types of ethylene copolymers disclosed) can be accomplished by several means. In one case, all ingredients (polymers, fillers, slip agents, anti-block, process aids, stabilizers, and the like.) in the reinforced polymer can be mixed together to form a complete polyethylene/filler mixture ready to be extruded into film, or alternatively, a "concentrate" of filler and/or additives can be made in one or all of the copolymers which is then "let down" or diluted with the balance of the ingredients and/or polymers to make the same polymer mixture. In the second case it is understood that sufficiently intensive mixing will occur in the film extruder to provide a homogeneous film.

It should be further understood that additives often found in films are contemplated by our invention as well. Such additives will be understood by those skilled in the art to include those that will have an effect on surface characteristics of films, processability of resins being made into films, thermal stability of resins or film and the like. These, and other types of additives, are normally carried in polyolefins, but may be added without such polyolefin carriers. The additive types mentioned are not meant to be a complete list, but merely illustrative. The additives included in the film will be understood to be different from the additives to be carried in compounding ingredient bags, while they may or may not differ in type, their intended use will differ.

The reinforcing fillers can be talc, preferably talc with a high aspect ratio, with or without surface treatment, or any other type of reinforcing filler including, but not limited to, the various forms of silica (fumed, precipitated, and the like)

with or without a coupling agent, other mineral type fillers including but not limited to other mineral type fillers such as kaolin and other clays, feldspar, silicas both natural and synthetic, carbide fillers, metallic oxides, sulfates, silicates and titanates, as well as carbon black. Such mineral type fillers may also include surface treatment discussed below and/or coupling agents. There are also other fillers that may enhance stiffness and tear resistance although the usual primary use of these fillers is to impact other properties such as electrical conductivity or flame retardancy. The surface treatment of talc can be stearic acid or any other common treatment intended to increase the affinity of the filler for the polyethylene. Aspect ratio of a platy filler such as talc is the platelet effective diameter to thickness ratio.  $\text{CaCO}_3$  is effectively a sphere and has an estimated aspect ratio of 1, while the talc contemplated in certain embodiments of my invention has an aspect ratio estimated in the range of 20-50. These filled films provide increased strength properties and stiffness which enable easier handling of wrapped/bagged product while still providing the dispersibility of a low melting film structure in a mechanical (rubber or elastomer) mixer.

In these preferred embodiments it is preferred that a reinforcing filler such as talc, and preferable talc with a high aspect ratio be included in the range of from 5 - 50%, and preferably in the range of from 10-40%, more preferably in the range of 15-30%, by weight based on the total weight of the polyethylene (or polyethylene blend) and the filler. Combinations of fillers are also contemplated. For example,  $\text{CaCO}_3$  and one or more of the talcs or other fillers described above are contemplated. Such combinations are part of contemplated embodiments as long as the final polyethylene/filler combination meets the desired melting, softening, and physical properties disclosed herein. The reinforcing filler level (wt%) will be based on the total weight of the polyethylene, or polyethylene blend, as well as the weight of any and all additives. The reinforcing filler should be present in the ethylene  $\alpha$ -olefin copolymer (or blends of ethylene  $\alpha$ -olefin copolymers) or in a blend of ethylene  $\alpha$ -olefin copolymer(s) with at least one ethylene copolymer in an amount effective to increase physical properties of the resulting film, while still maintaining a Vicat

softening point and DSC melting point within the described limits discussed earlier. That is the filler should be present to a level where the following film properties exists:

	Vicat		Melting Point	
	less than °C		less than °C	
	90		100	
preferred	80		90	
more preferred	70		80	
most preferred	60			

  

	Modulus		Tear	
	greater than °C		greater than °C	
	(K psi)		(g/ml)	
	MD	TD	MD	TD
	6	6	150	140
preferred	7	7	160	160
more preferred	8	8		200
most preferred	10	9		250

5

In the practice of embodiments of this invention, the rubber is generally compounded utilizing a mechanical mixer. The compounded rubber can include mixed unvulcanized rubber with the bale wrap and/or a compounding ingredient bag film of this invention. The compounded rubber can also include accelerators, promoters, curing or crosslinking agents, fillers, colorants, anti-oxidants, and other adjuvants.

10

While mono layer extrusion is exemplified, multilayer extrusion is also contemplated. One or more of the layers would have sufficient filler, to improve the physical properties of the resulting films. The base resin (polyethylene) or polyethylenes for each layer may be the same or different. In any case, an effective amount of filler must be in one or more layers to achieve improvement in one or more of film modulus, tear strength and the like.

15



Additionally, the polyethylene component of the eventual film may be as stated supra, a blend or coextrusion of one or more polyethylenes. For instance, a blend of 2 "ethylene  $\alpha$ -olefin copolymers" might include an ethylene, butene-1 copolymer and ethylene, octene-1 copolymer. Such blends could also include ethylene butene-1, 5 ethylene, hexene-1; ethylene, hexene-1, ethylene, octene-1; tri part blends such as ethylene, butene-1, ethylene hexene-1, and ethylene octene-1 are also contemplated.

Further, the so called "second ethylene copolymer" may be one or more ethylene copolymers such as ethylene vinyl acetate, ethylene methyl acrylate, ethylene ethyl acrylate, ethylene n-butyl acrylate, ethylene acrylic acid, ethylene methacrylic 10 acid, ionomers of the acid copolymers, terpolymers such as ethylene vinyl acetate, methylacrylate; ethylene methyl acrylate, acrylic acid and the like. The second ethylene copolymer may be also a blend of two or more such second ethylene copolymers.

The amount of second ethylene copolymer or copolymers blended optionally with an amount of the first ethylene copolymer or copolymers will be understood by 15 those of skill in the art, to be an effective amount to improve the processability of the first ethylene copolymer, generally with respect to bubble stability (blown films) or neck in (cast films). The use of second ethylene copolymers may also be to add other properties to the final film such as lower cost.

It might also be possible to use only the first ethylene copolymer without the 20 processing benefit of the second ethylene copolymer, especially considering the improvement in rheology by the use of fillers.

In any case when speaking of a "polyethylene/filler" blend film or composition, it will be understood that I intend the term polyethylene to include any such blends as discussed above or such blends that still provide a low melting and/or softening point. 25 The amount of filler (e.g. 5-50 weight %) will be based on the total weight of the polyethylene or polyethylene blend.

### EXAMPLES

#### Example 1

A film was made on a Sterling blown film extrusion line made by having a 1.5 30 inch diameter, 24:1 L/D screw and a 2 inch diameter die. The resin (polyethylene) blend for the film was made by dry blending pellets of 87.5 wt. % Exact® 4011

manufactured by Exxon Chemical Co. and 12.5 wt. % of a master batch (all ingredients shown below the Exact in the following formulation). This resulted in a final blend composition of:

		<u>Weight %</u>
5	Exact® 4011	87.50
	LD 760.36	10.24
	Erucamide	0.375
	Stearamide	0.375
	Diamataceous Earth	1.500
10	Dynamar® FX 9613	<u>.009</u>
		99.999 %

The Exact 4011 and LD 760.36 are available from Exxon Chemical Company. Exact 4011 is a butene ethylene copolymer with a nominal density of 0.885 g/cc and a 2.2 g/10 min. melt index. LD 760.36 is an EVA copolymer containing 27.5% vinyl acetate with a 2.3 g/10 min. melt index.

Dynamar® FX-9613 is a fluoropolymer from 3M Co. used to help eliminate melt fracture.

#### Example 2

A film was prepared on the same extrusion line as Example 1 using a polymer mixture reinforced with calcium carbonate, Hi Phlex® 100, from Specialty Minerals Corp. This mixture was prepared on a Werner Pfleider twin screw mixer.

		<u>Weight %</u>
	Exact 4011	64.5
	LD 760.36	10.0
25	Erucamide	0.07
	Dynamar FX 9613	0.14
	Ciba Geigy B-900	0.26
	Hi Phlex 100	<u>25.00</u>
		99.97 %

#### Example 3

A 10% talc mixture was prepared by dry blending the following and mixing in the same laboratory extruder as example 1. The 30% UTMT 609BF Concentrate used here was prepared by Specialty Minerals from 30% UTMT 609BF in Exact 4011 in their laboratory mixer. UTMT609BF is a treated talc having a high aspect ratio.

	<u>Weight %</u>
Exact 4011	54.5
30% UTMT 609BF Concentrate	33.0
SSABC0612VA	<u>12.5</u>
	100.0 %

This resulted in a film composition as follows:

	<u>Weight %</u>
Exact 4011	77.6
UTMT609BF	9.9
LD 760.36	10.24
Erucamide	.375
Stearamide	.375
Diatomaceous Earth	1.500
Dynmar FX 9613	<u>.009</u>
	99.999

#### Example 4

A 20% talc mixture was prepared in a similar manner as Example 3 from the same concentrate containing 30% treated talc (30% UTMT-609BF) in Exact 4011, specially prepared by Specialty Minerals in a laboratory mixer). This dry blend was let down in the same laboratory extruder to provide a 20% overall talc containing film.

	<u>Weight %</u>
Exact 4011	20.5%
30% UTMT 609BF Concentrate	67.0
SSABC0612VA	<u>12.5</u>
	100 %

resulting in a film composition of:

	<u>Weight %</u>
Exact 4011	67.4
UTMT 609BF	20.1
LD 760.36	10.24
Erucamide	0.375
Stearamide	0.375
Dia. Earth	1.500
Dynamar FX 9613	<u>.009</u>
	99.999

#### Example 5

A 26% talc mixture was prepared in a similar way by dry blending the following

	<u>Weight %</u>
30% UTMT 609BF Concentrate	87.5
SSABC0612VA	<u>12.5</u>
	100.0%

resulting in:

	<u>Weight %</u>
Exact 4011	61.2
UTMT609BF	26.3
LD 760.36	10.24
Erucamide	.375
Stearamide	.375
Diatomaceous. Earth	1.500
Dynamar FX 9613	.009
	99.999%

Prospective Examples 6, 7 and 8

Examples 1, 4 and 5 are repeated but the Exact 4011 is replaced by LD 760.36. It is expected that the inclusion of 20 and 25 weight percent talc (Examples 7 and 8) will not raise the Vicat softening point substantially above the 51°C of the resin itself. Further, at least a 30% improvement in secant modulus (MD & TD) is expected as well as a potential improvement in Elmendorf tear.

TABLE I

Example	1	2	3	4	5	Prospective Examples		
						6	7	8
Filler	None	CaCO <sub>3</sub>	Talc	Talc	Talc	None	Talc	Talc
Wt%	0	25%	10%	20.1%	26.3%	0	20%	25%
Gauge, mils	1.73	2.10	2.0	1.9	1.95	2.0	2.0	2.0
Secant Modulus, psi								
MD	6590	8630	8063	12400	13080			
TD	3500	4870	9197	10870	11870			
Elmendorf Tear, grams								
MD	229	356	253	539	337			
(g/mil)	132	169.5	126.5	283.6	172.8			
TD	218	386	305	607	606			
(g/mil)	126	183.8	152.5	319.5	310.7			
Power, amps	33	35	30	25	23			
Bubble Stability	least					most		
DSC Melting Point, °C	71.5	72.5	72.7	72.5	73.5			
Vicat Softening Point, °C	56.4	56.4	55.1	53.7	49.4	51	<55	<55

Table I shows the unexpected and surprising change in physical properties of Examples 1 through 5 as reinforcing filler is added. The stiffness and tear strength of Example 2 using calcium carbonate is improved over Example 1, while the DSC melt point has not been substantially increased, and the Vicat has not changed in spite of the significant reinforcement. The use of talc in Examples 3, 4, and 5 surprisingly and unexpectedly provides even greater reinforcement as shown by the large increase in stiffness (secant modulus) and several times increase in tear resistance. In addition, the processing characteristics improved as reinforcing filler was added as shown by a decrease in power consumption and an improvement in bubble stability. Very important is the decrease of Vicat softening point temperature as talc content is increased indicating an increase in ease of dispersability while increasing the toughness of the matrix.

#### Conclusion

The present invention has been described in considerable detail with reference to certain preferred versions thereof, other versions are possible. For example, while rubber bale wrap and compounding ingredient bag films from polyethylene talc combinations have been exemplified, other uses are also contemplated. Therefore, the spirit and scope of the appended claims should not be limited to the description of the preferred versions contained herein.

**CLAIMS**

I claim:

1. A batch inclusion bag film having a Vicat softening point below 90° C, a melting point below 100° C, a 1% secant modulus (MD) above 10,000 psi and (TD) above 6000 psi, and an Elmendorf tear (g/mil) above 150 (MD), 200 (TD), comprising:
  - a) at least a first ethylene  $\alpha$ -olefin copolymer, said first copolymer having a density below 0.915 g/cc; and
  - b) in the range of from 5-50 % by weight of talc, said weight percent based on the total of the ethylene  $\alpha$ -olefin copolymer.
2. The batch inclusion bag of claim 1 wherein said film has a Vicat softening point less than 80 °C, preferably less than 70 °C, more preferably less than 60 °C, a melting point below 90°C, preferably below 80°C, more preferably below 80°C, a 1 % secant modulus exceeding 7,000 psi (MD) and (TD), preferably exceeding 8000 psi (MD) and (TD), more preferably exceeding 10,000 psi (MD) and 9000 psi (TD) and an Elmendorf tear above 160 g/mil (MD) and (TD), preferably above 200 g/mil, more preferably above 250 g/mil.
3. The batch inclusion bag of any of the preceding claims wherein said ethylene  $\alpha$ -olefin copolymer is one of ethylene, 1-butene; ethylene, 1-hexene; ethylene, 1-octene; or combinations thereof, said copolymer having a density below 0.910 g/cc; wherein said talc is one of talc, high aspect ratio talc, surface treated talc, surface treated high aspect ratio talc, or combinations thereof, and is present in said film in the range of 10-40 weight percent, preferably 15-30 weight percent, based on the total weight of said ethylene  $\alpha$ -olefin copolymer.

4. The batch inclusion bag of any of the preceding claims, wherein said film further includes an ethylene ethylinically unsaturated carboxylic acid ester copolymer.
5. The batch inclusion bag of any of the preceding claims, wherein said film further includes calcium carbonate.
6. A package comprising:
- a) an article being one of unvulcanized rubber, vulcanized rubber, rubber compounding ingredients, or combinations thereof, wherein said article is covered and in contact with a film;
- b) said film having a Vicat softening point below 70° C, a 1% secant modulus (MD & TD) above 8,000 psi, an Elmendorf tear of at least 150 g/mil (MD), 200 g/mil (TD), said film comprising:
- i) at least a first ethylene  $\alpha$ -olefin copolymer of ethylene and one or more alpha-olefins, said alpha-olefins being one of butene-1, hexene-1, octene-1 or combinations thereof, having a density below 0.910 g/cc, and
- ii) a filler, said filler being present in said film at a level of at least 10 weight percent based on the total weight of said ethylene copolymer.
7. The package of claim 6, wherein said filler is one of talc, surface treated talc, high aspect ratio talc, high aspect ratio surface treated talc, clays, feldspar, silicas, carbides, metallic oxides, sulfates, silicates, titanates, or combinations thereof.
8. The package of claim 6, wherein said filler is one of talc, surface treated talc, high aspect ratio talc, high aspect ratio surface treated talc, or combinations thereof.

9. The package of claim 6, wherein said film comprises:
- a) a polyethylene or polyethylene blend; and
  - b) a effective amount of a filler to impart to said film a secant modulus of at least 10,000 psi (MD) and 9,000 psi (TD), and an Elmendorf tear of at least 160 g/mil (MD) and 200 g/mil (TD).
10. The package of claim 8 wherein said polyethylene is an ethylene, alpha-olefin copolymer wherein said alpha-olefin is one of propylene, butene-1, pentene-1, 4-methyl-1-pentene, hexene-1, octene-1, or combinations thereof, said copolymer having a density below 0.910 g/cc.
11. The package of claim 8 wherein said polyethylene further includes a second ethylene copolymer, wherein a comonomer in said second ethylene copolymer is one of vinyl acetate, methyl acrylate, ethyl acrylate, n-butyl acrylate, acrylic acid, methacrylic acid or combinations thereof. wherein said second ethylene copolymer is present in the range of 1 to 25 weight percent based on the total weight of ethylene copolymers.
12. The package of claim 8 wherein said filler is one of talc, surface treated talc, high aspect ratio talc, high aspect ratio surface treated talc, or combinations thereof.



## INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 97/22069

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08J5/18 C08K3/34 C08L23/08 //C08L23:08

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08J C08K C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 742 248 A (MITSUI PETROCHEMICAL IND) 13 November 1996 see the whole document ---	1-12
A	WO 95 07314 A (DOW CHEMICAL CO) 16 March 1995 cited in the application see the whole document ---	1-12
A	WO 95 09199 A (EXXON CHEMICAL PATENTS INC) 6 April 1995 see the whole document & US 5 500 260 A cited in the application ----- -/--	1-12

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents:

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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 385 599 A (EXXON CHEMICAL PATENTS INC) 5 September 1990 see the whole document & US 5 145 747 A cited in the application ---	1-12
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Information on patent family members

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